Non-linear far-infrared spectroscopy with an FEL

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Outline

- Nonlinear optics in the THz frequency range
- Nonlinear susceptibility of III-V semiconductors
- Investigation of the nonlinear susceptibility of GaAs below the Reststrahlen band with an FEL
- Conclusion

Nonlinear optics in the THz frequency range I

- Nonlinear optics (frequency mixing, harmonic generation,...) well established in the wavelength range > 20 µm to VIS/UV
- In the THz frequency range (1-10 THz, 300-30 µm) no convenient, high power light sources are available for NLO ("THz gap")
- NLO experiments at THz frequencies are based on
  - frequency mixing of visible with FIR gas laser/microwaves [1)]
  - FIR gas lasers operating at discrete frequencies [2)]
  - tunable free-electron lasers
- Recent developments:
  - THz quantum cascade lasers [3)]
  - DFG/OR with intense fs pulses [4)]
  - DFG coherent synchrotron THz radiation [5)]

1) Faust and Henry, PRL **17**, 1265 (1966).
2) Mayer, Keilmann, PRB **33**, 6954 (1986), review: Keilmann, Infrared Phys. **31**, 373 (1991).
3) Köhler et al., Nature **417**, 156 (2002).
4) e.g. Reimann et al., Optics Lett. **28**, 471 (2003).
5) Carr et al., Nature **420**, 153 (2002).

Nonlinear optics in the THz frequency range II

- What is interesting in NLO at THz frequencies?
  - Many elementary low-energy excitations lie in the THz frequency range, e.g. phonon-polaritons, plasmons, gap energies of superconductors, collective vibrations of biomolecules etc...
  - Their coupling to radiation gives rise to resonant enhancement of NLO processes
  - Possibility to obtain information on nonlinear susceptibility at resonances
- Here: investigation of second harmonic generation below the optical phonon resonance in GaAs[1)].
  - detailed information on nonlinear terms of the lattice potential

1) Dekorsy et al. Phys. Rev. Lett. **90**, 055508 (2003).

Theory: 2nd order nonlinear susceptibility

- General description of $\chi^{(2)}$ in the vicinity of a phonon-polariton resonance[1)]:

$$\chi^{(2)}(\omega_3,\omega_2,\omega_1) = \chi_E^{(2)}\left[1 + C_1\left(\frac{1}{D(\omega_1)} + \frac{1}{D(\omega_2)} + \frac{1}{D(\omega_3)}\right) + C_2\left(\frac{1}{D(\omega_1)D(\omega_2)} + \frac{1}{D(\omega_1)D(\omega_3)} + \frac{1}{D(\omega_2)D(\omega_3)}\right)\right.$$
$$\left. + C_3\left(\frac{1}{D(\omega_1)D(\omega_2)D(\omega_3)}\right)\right]$$

with $$D(\omega) = 1 - \left(\frac{\omega}{\omega_{TO}}\right)^2 - \frac{i\omega\gamma}{\omega_{TO}^2}$$

$\chi_E^{(2)}$: pure electronic part

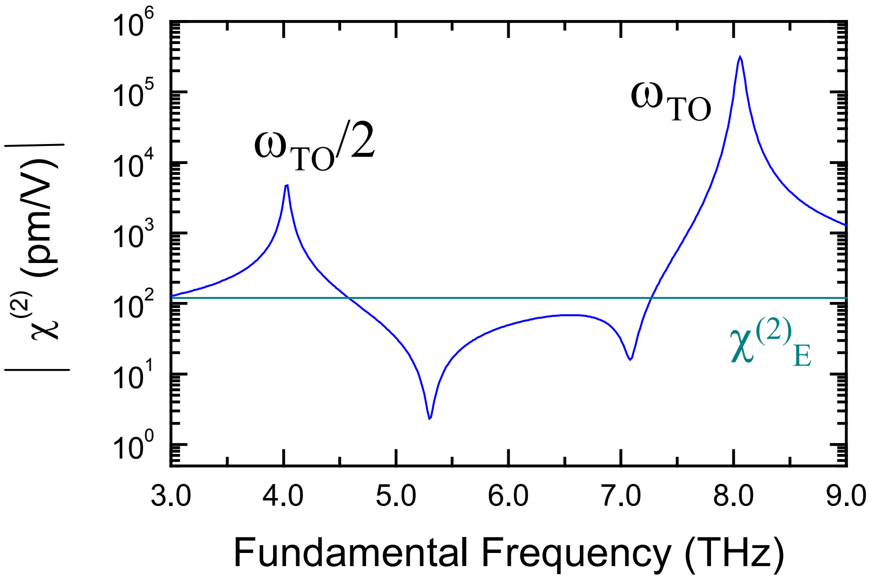
ω_{TO} : TO phonon frequency

γ : phonon daming constant

C_1: Faust-Henry coefficient (Pockels effect, frequency mixing, Raman I_{TO}/I_{LO})
C_2: second-order lattice dipole moment
C_3: third-order lattice potential anharmonicity

1) Flytzanis, Phys. Rev. B **6**, 1264 (1972).

$\chi^{(2)}(\omega_3=2\omega_1)$ of GaAs in the THz range



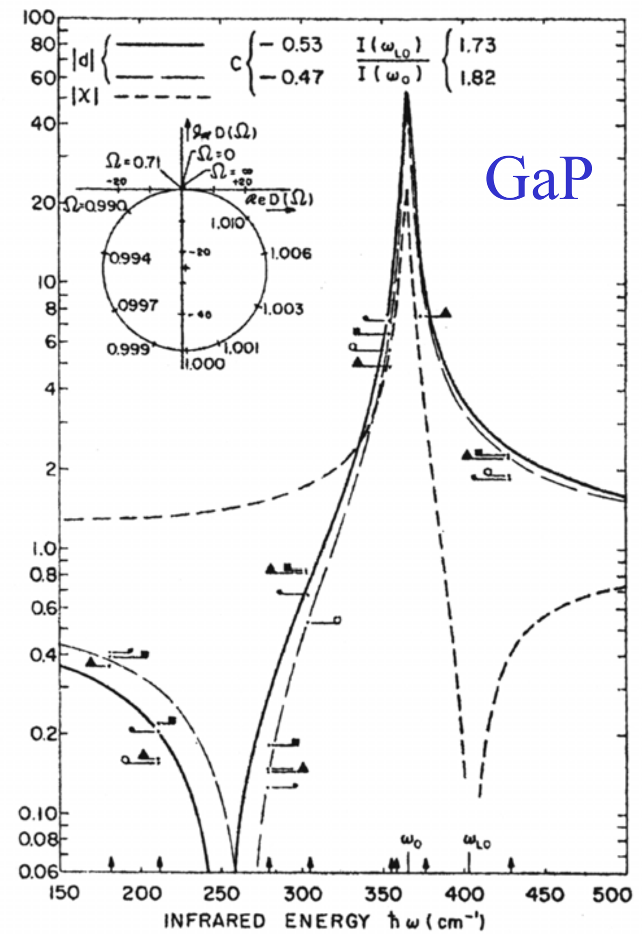
- ω_{TO}=8.05 THz
- $\chi^{(2)}_E$=132 pm/V
- C_1= -0.59 (Faust-Henry coeff.)
- C_2,C_3 not accurately known: theory: C_2 =0.14, C_3=-0.07, $3C_2+C_3$=0.35; experiment $3C_2+C_3$=0.39

- strong resonance at ω_{TO}, exceeding $\chi^{(2)}_E$ by factor of 2100
  - observed before in GaP[1)]
- resonance at $\omega_{TO}/2$, exceeding $\chi^{(2)}_E$ by a factor of 33
- 2 minima in $\chi^{(2)}$ between $\omega_{TO}/2$ and ω_{TO}
  - resonance and minima not observed before
  - observation would allow determination of C_i's without need for determination of absolute intensities

1) Barmentlo et al. Phys. Rev. A **50**, R14 (1994)

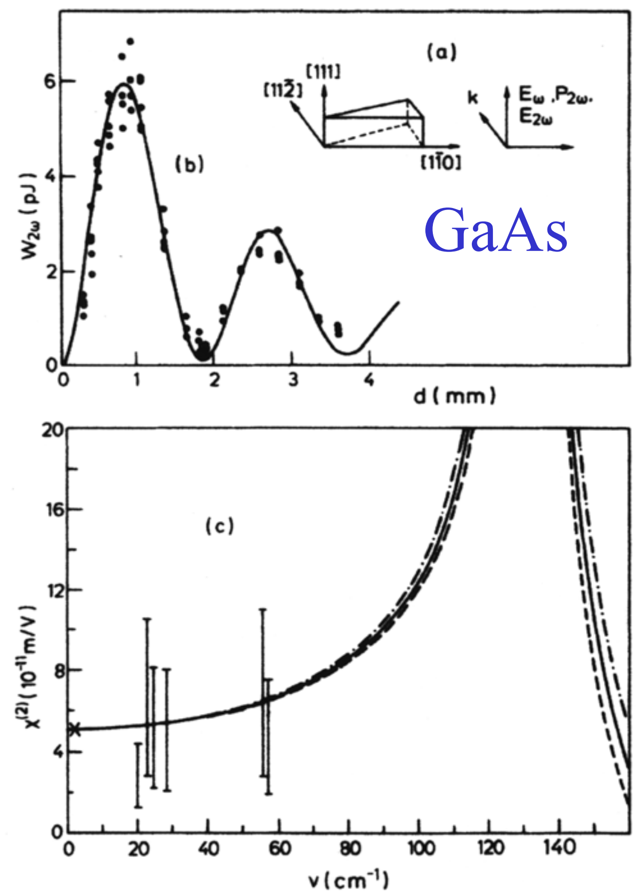
Previous nonlinear optical determinations of C_i's

frequency mixing of 632 nm + (20-55) µm



Faust et al., PR **173**, 781 (1968)

SHG with CH_3F laser (175-500 µm)

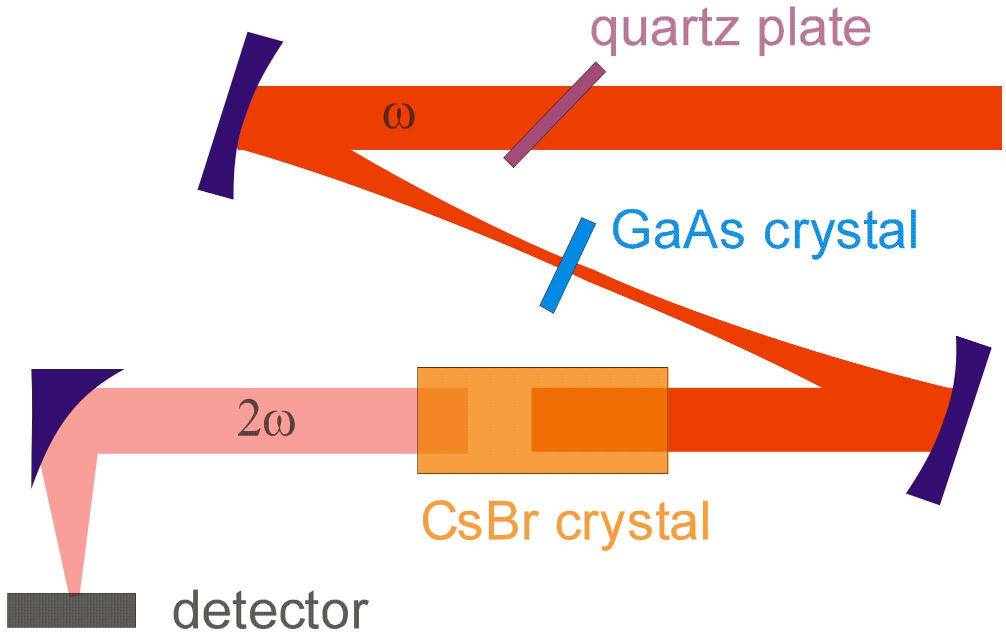


Mayer & Keilmann, PRB **33**, 6954 (1986)

Measurement of $\chi^{(2)}$ with a free-electron laser

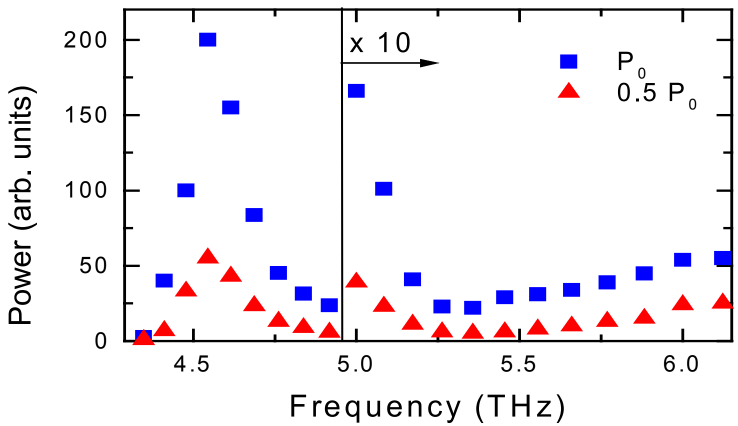
- free-electron laser FELIX, Rijnhuizen, NL
- tunable in the THz frequency range (1.2 THz-70 THz)
- macro-pulses with 10 Hz repetition rate with 100 micro-pulses at 25 MHz
- picosecond pulse width, 0.2 THz spectral width
- 4 - 8 µJ energy per pulse, average power 40-80 mW
- focused on ~ 500 µm spot $\Rightarrow$ 30 kV/cm electric field

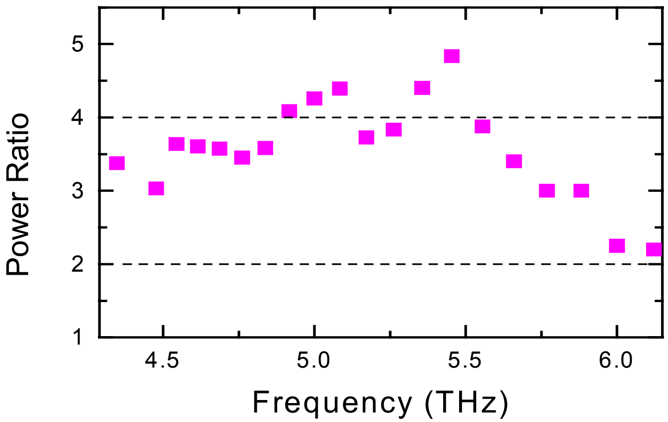
Experimental set-up



- 2.7 mm quartz plate: cleans FEL pulses from higher harmonics (fundamental > 4.3 THz) better than 3.5×10^{-7}
- 2×2.7 cm CsBr crystal: blocks FEL fundamental before detector between 4 THz to 5.5 THz better than 10^{-19}, losses for SHG of 66 %
- high sensitivity helium cooled Ge:Ga detector
- whole set-up evacuated

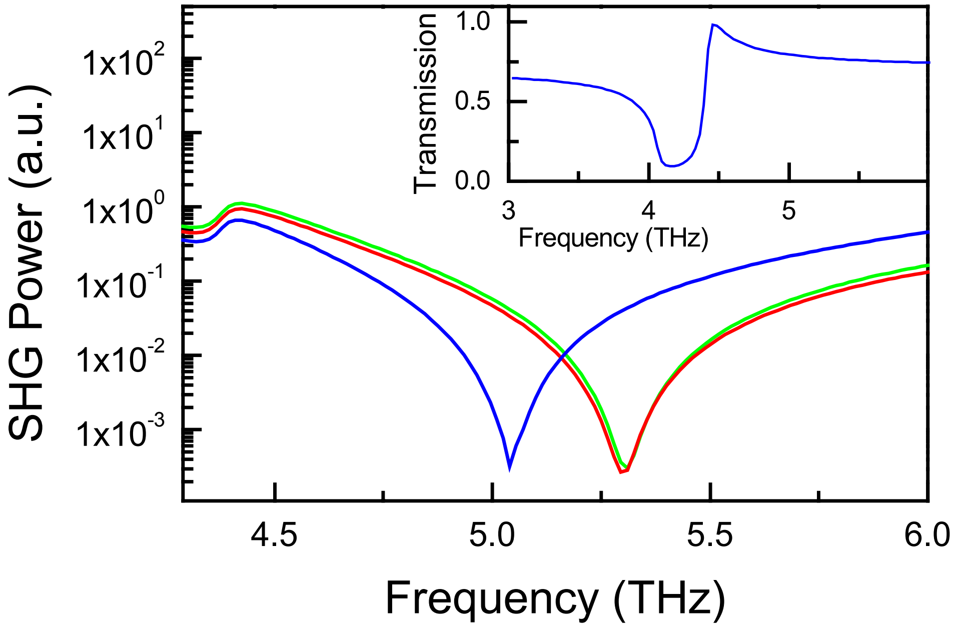
Experimental result on 18 µm thick (211) oriented GaAs crystal





- Detection of SHG power for two different incident powers
- Power ratio confirms sensitivity of set-up to SHG between 4.3 THz to 5.6 THz
- Strong maximum at 4.5 THz (>$\omega_{TO}/2$=4.0 THz)
- Minimum at 5.3 THz

Theoretical modeling of observed zero-crossing frequency

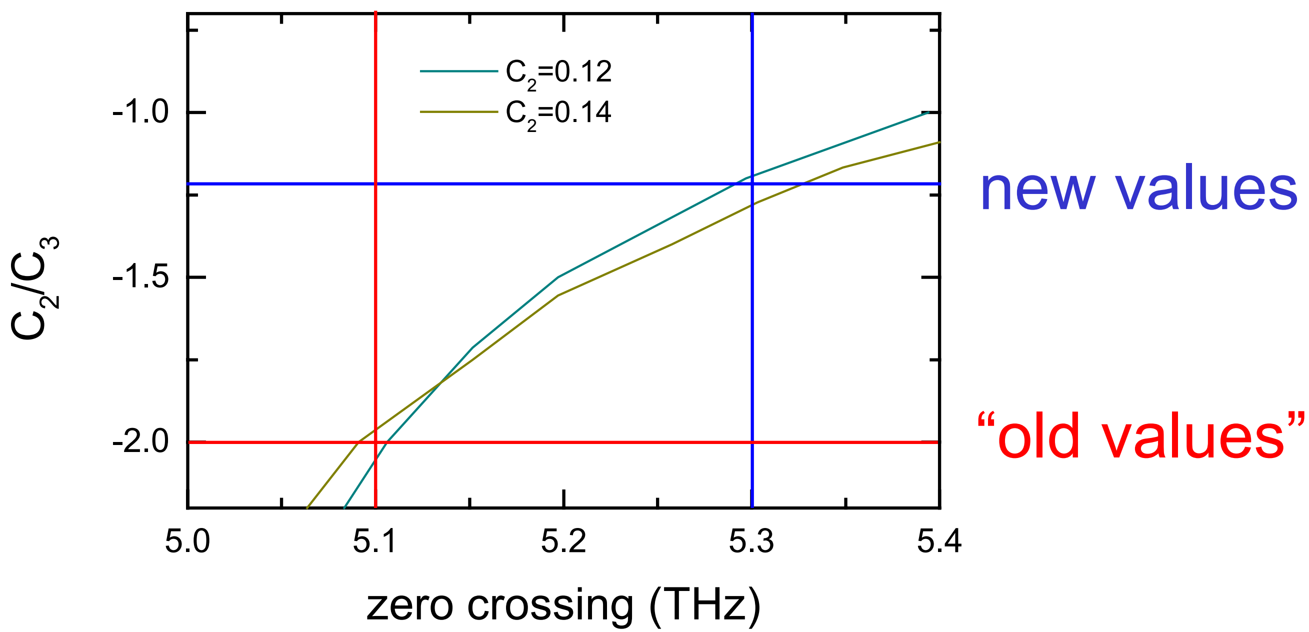


$3C_2+C_3=0.35$, $C_2/C_3= -2.0$
(old theoretical values)

$3C_2+C_3=0.35$, $C_2/C_3= -1.23$
$3C_2+C_3=0.39$, $C_2/C_3=- 1.3$
(fits to our experiment)

- calculation of SHG considering coherence length, absorption, Fresnel coefficients for outcoupling
- observed zero-crossing point of $\chi^{(2)}$: contribution from phonon interaction through the third-order lattice potential anharmonicity vs second order lattice dipole moment is significantly larger than calculated by theory

Uncertainty of determined C_i's



- error in determination of zero-crossing of $\chi^{(2)} < 0.1$ THz
- ambiguity in determination of C_2 and C_3
- but: ratio of C_2/C_3 has to be significantly changed

Conclusion

- Investigation of dispersion of second order nonlinear susceptibility in GaAs with an free-electron laser
- First observation of resonance close to half the phonon frequency (4.5 THz)
- Determination of zero-crossing point of $\chi^{(2)}$ at 5.3 THz
- New values for higher order lattice potentials to the nonlinear susceptibility: contribution from phonon interaction through the third-order lattice potential anharmonicity vs second order lattice dipole moment is significantly larger than calculated by theory[1)]
- New method for determination of these parameters in other materials

1) Dekorsy et al. Phys. Rev. Lett. **90**, 055508 (2003).

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